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CALCULATIONS OF COLLISION-INDUCED OPTICAL GAIN IN
HIGH-DENSITY MIXTURES OF D2 OR HD WITH A BUFFER GAS(U)
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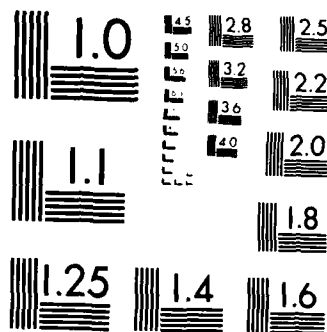
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MELBOURNE, VICTORIA

REPORT

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**CALCULATIONS OF COLLISION-INDUCED OPTICAL GAIN IN
 HIGH-DENSITY MIXTURES OF D₂ OR HD
 WITH A BUFFER GAS**

R. McLeary

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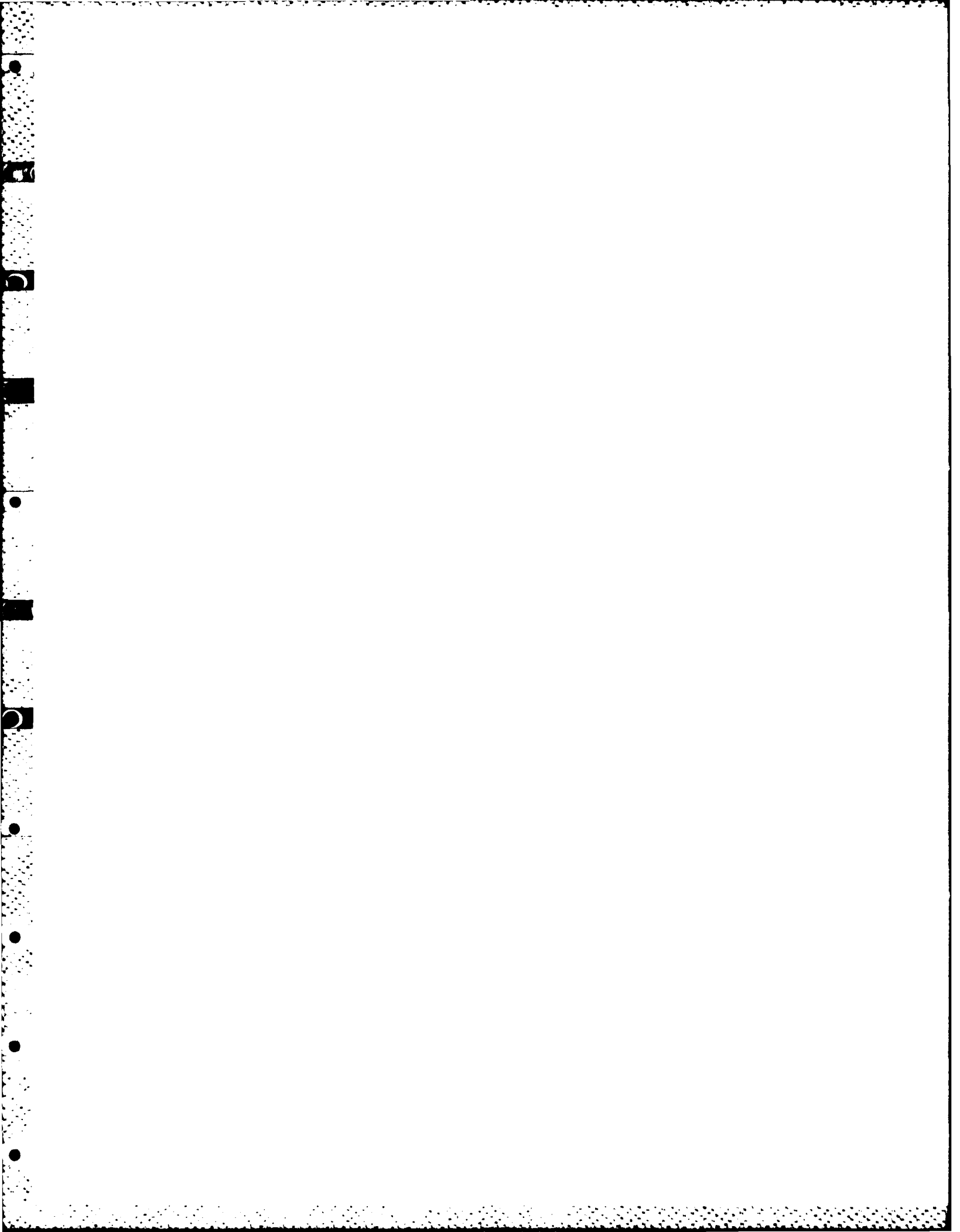


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ABSTRACT

This report presents the results of calculations of collision-induced optical gain in vibrationally-excited mixtures of deuterium or hydrogen deuteride with a buffer gas. Gains of up to 0.07 cm^{-1} are predicted for gas mixtures at high pressure and reduced temperature. Significant gain is predicted over a frequency range of approximately 2100 cm^{-1} to 2700 cm^{-1} (wavelength range $4.8 \text{ } \mu\text{m}$ to $3.7 \text{ } \mu\text{m}$) for D_2 mixtures and over the frequency range 2600 cm^{-1} to 3300 cm^{-1} (wavelength range $3.8 \text{ } \mu\text{m}$ to $3.0 \text{ } \mu\text{m}$) for HD mixtures. These results indicate the feasibility of operation of collision-induced-dipole lasers which are continuously tunable over the above frequency ranges.

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ABSTRACT

This report presents the results of calculations of collision-induced optical gain in vibrationally-excited mixtures of deuterium or hydrogen deuteride with a buffer gas. Gains of up to 0.07/cm⁻¹ are predicted for gas mixtures at high pressure and reduced temperature. Significant gain is predicted over a frequency range of approximately 2100/cm⁻¹ to 2700/cm⁻¹ (wavelength range 4.8 μm to 3.7 μm) for D₂ mixtures and over the frequency range 2600/cm⁻¹ to 3300 cm⁻¹ (wavelength range 3.8 μm to 3.0 μm) for HD mixtures. These results indicate the feasibility of operation of collision-induced-dipole lasers which are continuously tunable over the above frequency ranges.

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CALCULATIONS OF COLLISION-INDUCED OPTICAL GAIN IN
HIGH-DENSITY MIXTURES OF D₂ OR HD WITH A BUFFER GAS

1. INTRODUCTION

This report presents the results of calculations of collision-induced optical gain in vibrationally-excited mixtures of D₂ or HD with a buffer gas. These two molecules have collision-induced-absorption profiles [1],[2] which encompass the output wavelengths of the hydrogen-fluoride (HF) laser. The HF laser therefore provides a ready means of vibrationally-exciting these molecules [3]. There is no equivalent high-power laser which has an output wavelength which lies within the absorption profile of H₂ [4]. Thus H₂ has not been analysed, even though its large rotational energy [5] is expected to result in gains of greater magnitude than those calculated for D₂ or HD.

The calculations presented here are similar to those carried out by Christiansen and Greenfield [6], who considered a vibrationally-excited mixture of H₂ and Xe at a total density of 15 Amagat*. In their analysis the gases were vibrationally-excited by an electric discharge. These authors predicted extremely low gains of approximately 10^{-4} cm^{-1} , a value which is unlikely to be achieved in practice when various absorption mechanisms not included in the analysis are present. In this report, optically-excited mixtures at much higher densities (~ 700 Amagat) are investigated, and gains of up to 0.07 cm^{-1} are predicted.

* One Amagat is the density at STP.

2. MECHANISMS FOR ACHIEVING GAIN

The measured fundamental-band absorption profile [1] for a mixture of D_2 and Ar is shown in Fig. 1. This figure shows only the contribution from D_2 :Ar collisions; there is an extra component due to D_2 : D_2 collisions which is much smaller and of slightly different profile [7], and which is not included in this figure. The absorption profile consists of the summation of a number of vibration-rotation transitions of the O, Q and S branches, where $\Delta J = -2, 0$ and $+2$ respectively, and J is the rotational quantum number in the ground vibrational state. The positions of the line centres of the strong transitions are shown on the frequency axis.

The absorption due to O-branch transitions from rotational levels with $J > 4$ is small, since these levels are sparsely populated at room temperature. When a population of vibrationally-excited molecules is established in the first excited vibrational state, most of the molecules similarly lie in levels of low rotational quantum number. For moderate levels of vibrational excitation ($\approx 10\%$) a population inversion is achievable in D_2 on O-branch transitions with $J > 3$.

A population inversion may similarly be achieved in HD. However, due to its larger rotational constant [5], inversion will be readily achieved when $J > 2$.

The line shapes $L(\nu)$ of these collision-induced-dipole transitions are not symmetric with respect to frequency (ν) about the line centre ($\bar{\nu}$). In absorption the high-frequency wing has a Lorentzian profile, while the low-frequency wing is cut off exponentially. For absorption, the relation between the low and high-frequency wings is [2]:

$$L_A(\bar{\nu} - \Delta\nu) = L_A(\bar{\nu} + \Delta\nu) \exp(-hc\Delta\nu/kT)$$

where $\Delta\nu$ is the frequency shift (cm^{-1}) from line centre, h is Planck's constant, k is Boltzmann's constant, c is the velocity of light, and T is the temperature. The reason for the asymmetry is that the colliding molecule can either donate or accept kinetic energy during the collision with the absorbing molecule. Its capacity to donate kinetic energy is limited and is determined by the temperature; hence the cut-off on the low-frequency wing in absorption. A similar argument predicts that the cut-off will be on the high-frequency wing in emission, although this has not been confirmed experimentally.

This difference between the line shapes in absorption and emission enhances the gain on the low-frequency wings of the O-branch transitions. It can even lead to predictions of gain on the low-frequency wings of Q-branch and S-branch transitions. However these are small effects [6] except at low temperatures and Q and S-branch transitions are not considered in this report.

Another possible contribution to gain at frequencies on the low-frequency side of the absorption profile results from collisions between an excited diatomic molecule and an unexcited diatomic molecule. For this type of collision a "double" transition can occur, where the excited molecule makes a pure vibrational transition ($\Delta J = 0$), and the colliding molecule makes a pure rotational transition ($\Delta J = 2$). The probability of these transitions is considered to be relatively large at high density [4]. An estimate for the contribution of these transitions to the gain cannot be derived in the same way as is presented in this report for the O-branch transitions. For this reason, the double transitions are not further considered, although there is some evidence [8] to suggest that they can make a significant extra contribution to the gain.

3. GAIN CALCULATIONS

In the following, gain is calculated only for O-branch transitions between the ground and first vibrational level. This vibrational transition is expected to be the main contributor to the gain for the relatively low levels of excitation considered. The gain $\Gamma(J)$ for a transition with rotational quantum number J in the ground vibrational level, is given by:

$$\Gamma(J) \propto N \left\{ \frac{\epsilon N_{v'J'} L_E}{N g_{J'}} - \frac{(1-\epsilon) N_{vJ} L_A}{N g_J} \right\} \bar{\nu}_J S_J \langle vJ | Q | v'J' \rangle^2$$

where:

- ϵ = fraction of molecules in the first vibrational state.
- $v' (=1), v (=0)$ = vibrational quantum numbers.
- J' = $J-2$ i.e. O-branch transitions.
- $\bar{\nu}_J$ = frequency of transition (line centre).
- L_A, L_E = line shape functions in absorption and emission respectively.
- N = number density of diatomic molecules.
- $\epsilon N_{v'J'}$ = number density of diatomic molecules in vibrational level v' , rotational level J' .
- $(1-\epsilon) N_{vJ}$ = number density of diatomic molecules in vibrational level v , rotational level J .
- $g_J, g_{J'}$ = degeneracy factors.
- S_J = $J(J-1)/(2J-1)$.
- $\langle vJ | Q | v'J' \rangle$ = quadrupole matrix element.

Values for $N_{v'J'}/(Ng_{J'})$ and $N_{vJ}/(Ng_J)$, $\bar{v}_{J'}$, L_A and L_E , and $\langle vJ|Q|v'J' \rangle$ for mixtures of HD or D₂ with a buffer gas are given in appendices A, B, C, and D.

The absorption of the O(2) line at line centre in D₂ and HD can be found from experimental absorption profiles presented in references [1] and [2]. For D₂ mixtures, the absorption of the O(4) line can be found directly and the absorption of the O(2) line calculated from the O(4) value. Values at other densities and mixture ratios can be derived using scaling factors presented in [1] and [2]. In this report the gain $\Gamma(J)$ will be calculated in terms of these known absorptions which are designated A(2). The absorption of the O(2) line has been chosen as it is only a slowly varying function of the temperature, and thus approximately constant, over the temperature region of interest (see discussion section of this report). Thus:

$$\frac{\Gamma(J)}{A(2)} = - \frac{\left\{ \frac{\epsilon N_{v'J'} L_E}{Ng_{J'}} - \frac{(1-\epsilon) N_{vJ} L_A}{Ng_J} \right\} \bar{v}_{J'} S_J \langle vJ|Q|v'J' \rangle^2}{\frac{N_{v2}}{Ng_2} \bar{v}_2 S_2 \langle v2|Q|v'O \rangle^2} \quad (1)$$

where the values of the parameters appropriate to the particular diatomic molecule are used in the above expression.

The expression given in (1) is evaluated for a number of values of the excitation fraction ϵ and the temperature T, for J values 2, 3, 7. The calculated individual absorption/gain lines are then summed to give the total O-branch absorption/gain profile as a function of frequency. Positive values indicate absorption whilst negative values indicate gain.

4. RESULTS

Results of calculations of the O-branch absorption/gain for D₂ or HD mixed with a buffer gas (Ar or Kr) are presented in Figures 2-15. Normalised results are presented for excitation percentages (ϵ) of 0%, 5% and 10% at temperatures of 300 K, 220 K and 160 K. Values of 220 K and 160 K are approximately 10 K above the critical temperatures of Kr and Ar respectively.

Figure 2 shows the calculated O-branch normalised absorption profile ($\epsilon=0$) at 300 K for the D₂ mixture. The strong component transitions of the profile are shown as dashed curves on the figure. The J value is indicated on each transition.

Figure 3 shows the O-branch absorption/gain profiles at 300 K for 0%, 5% and 10% of the D₂ molecules excited to the first vibrational state. Figures 4 and 5 are similar results obtained at temperatures of 220 K and

160 K. Figures 6, 7 and 8 are the same results as presented in Figures 3, 4 and 5, but grouped by temperature rather than by excitation percentage.

Figures 9-15 are results for HD mixtures which correspond to the D_2 results presented in Figures 2-8.

5. DISCUSSION

In order to obtain absolute values of the gain for a particular gas mixture and temperature, the appropriate value of the normalising factor $A(2)$ has to be obtained. An estimate for $A(2)$, and hence the gain, for a number of conditions of interest is derived in this section.

The experimental curve of Figure 1 [1] indicates that at a frequency of 2573 cm^{-1} corresponding to the line centre of the $O(4)$ transition, the absorption is 0.008 cm^{-1} . This absorption is for a D_2 :Ar mixture of 95 Amagat D_2 and 230 Amagat Ar at room temperature. Since the wings of Q and S branch transitions make only a small contribution at this frequency, the value of 0.008 cm^{-1} corresponds to absorption due to O-branch transitions, primarily the $O(4)$ transition. The magnitude of the $O(4)$ absorption at line centre is approximately 80% of the total O-branch contribution (Figure 2), or 0.0064 cm^{-1} . The absorption of the $O(2)$ transition at line centre has a magnitude of 2.7 times that of the $O(4)$ line (Figure 2) and thus a value of 0.017 cm^{-1} . This value can be scaled to other gas mixtures and densities by means of data presented in [1]. For example, a 1:2 mixture of D_2 and Ar at room temperature and total density 730 Amagat has an $O(2)$ -line absorption ($A(2)$) of 0.1 cm^{-1} . A density of 730 Amagat at room temperature requires a pressure of 200 MPa and is readily achieved in practice [3],[8].

At constant density but reduced temperature, the collision-induced dipole effect (e.g. [13]) and the fraction of molecules in the $J=2$ rotational level (Appendix A) are both reduced. These will lower the value of $A(2)$. On the other hand if the pressure is kept constant as the temperature is reduced, the gas density increases and a larger induced-dipole effect results. The net result is that for D_2 :Ar mixtures at constant pressure, the value of $A(2)$ is approximately independent of temperature over the range of interest. Thus a value of 0.1 cm^{-1} for the normalising factor $A(2)$ is appropriate for a 1:2 mixture of D_2 and Ar at a pressure of 200 MPa for the three temperatures considered in this work.

If Ar is replaced by Kr at the same density, larger induced-dipole effects are anticipated [2], [12]. However, since Kr is a larger molecule than Ar, it is not as compressible, lower densities result, and this tends to cancel out its advantage as a collision partner. Thus the value 0.1 cm^{-1} calculated for the normalising factor of a 1:2 D_2 :Ar mixture at 200 MPa is also approximately correct for a similar D_2 :Kr mixture at the same pressure.

The normalising factor for HD mixtures can be obtained in a similar fashion using data presented in [2] and [12]. It turns out that a value of approximately 0.1 cm^{-1} also applies to a HD mixture (1:2 at 200 MPa) at temperatures of 300 K and 220 K. For a temperature of 160 K a value of 0.08 cm^{-1} is a better estimate; the lower value arises due to the greater dependence on temperature of the population in the $J=2$ rotational level for HD compared with D_2 (Appendix A).

Thus for deuterium:buffer-gas mixtures (1:2) at a pressure of 200 MPa and an excitation (ϵ) of 10%, the maximum gains (Figure 8) at temperatures of 300 K, 220 K, and 160 K are 0.007 cm^{-1} , 0.016 cm^{-1} , and 0.029 cm^{-1} respectively. For hydrogen-deuteride mixtures under the same conditions, the corresponding values derived from Figure 15 are 0.021 cm^{-1} , 0.035 cm^{-1} , and 0.069 cm^{-1} .

Significant gain is predicted for D_2 over a frequency range from about 2100 cm^{-1} to 2700 cm^{-1} (Figure 8) and for HD over a range from 2600 cm^{-1} to 3300 cm^{-1} (Figure 15). Since D_2 absorbs heavily in the range 2600 cm^{-1} to 3300 cm^{-1} (Figure 1), gain would only be achieved in HD mixtures which have low concentrations of D_2 impurity.

Larger gains would be predicted for values of the excitation parameter (ϵ) greater than 10%. However, in practice it is difficult to obtain a large proportion of the diatomic molecules in only the first vibrational level. For large excitations, an analysis which considers the higher vibrational levels would be required.

6. CONCLUSION

Calculations have been made of collision-induced gain in high-density mixtures of vibrationally-excited D_2 or HD with a buffer gas. Maximum gains of approximately 0.03 cm^{-1} (D_2) and 0.07 cm^{-1} (HD) have been predicted in 1:2 mixtures at a temperature of 160 K and a total pressure of 200 MPa.

Experimental gain measurements made in a 1:2 mixture of D_2 and Ar at 200 MPa are reported in [8]. For the conditions of this experiment ($T=300 \text{ K}$, $\epsilon \approx 4\%$), a decrease of absorption of about 0.045 cm^{-1} was measured over the frequency range 2520 cm^{-1} to 2640 cm^{-1} . This is nearly three times the value (Figure 3) predicted by the analysis presented in this report. It is argued in [8] that the contribution from "double" collisions, together with uncertainty in the value of ϵ in the experiment may account for the discrepancy between the experimental results and these calculations.

The results presented for the gain in this report can therefore be considered as minimum values; depending on conditions, somewhat larger values can be expected in practice.

Gains of these magnitudes make feasible the operation of a collision-induced-dipole laser, which would have continuous tunability over a range of frequency. In practice, substantial gain over a frequency range of approximately 2100 cm^{-1} to 2700 cm^{-1} for D_2 and 2600 cm^{-1} to 3300 cm^{-1} for HD is expected. In addition, the large bandwidths of the transitions should allow efficient energy extraction in short (\sim picosecond) pulses.

7. ACKNOWLEDGEMENT

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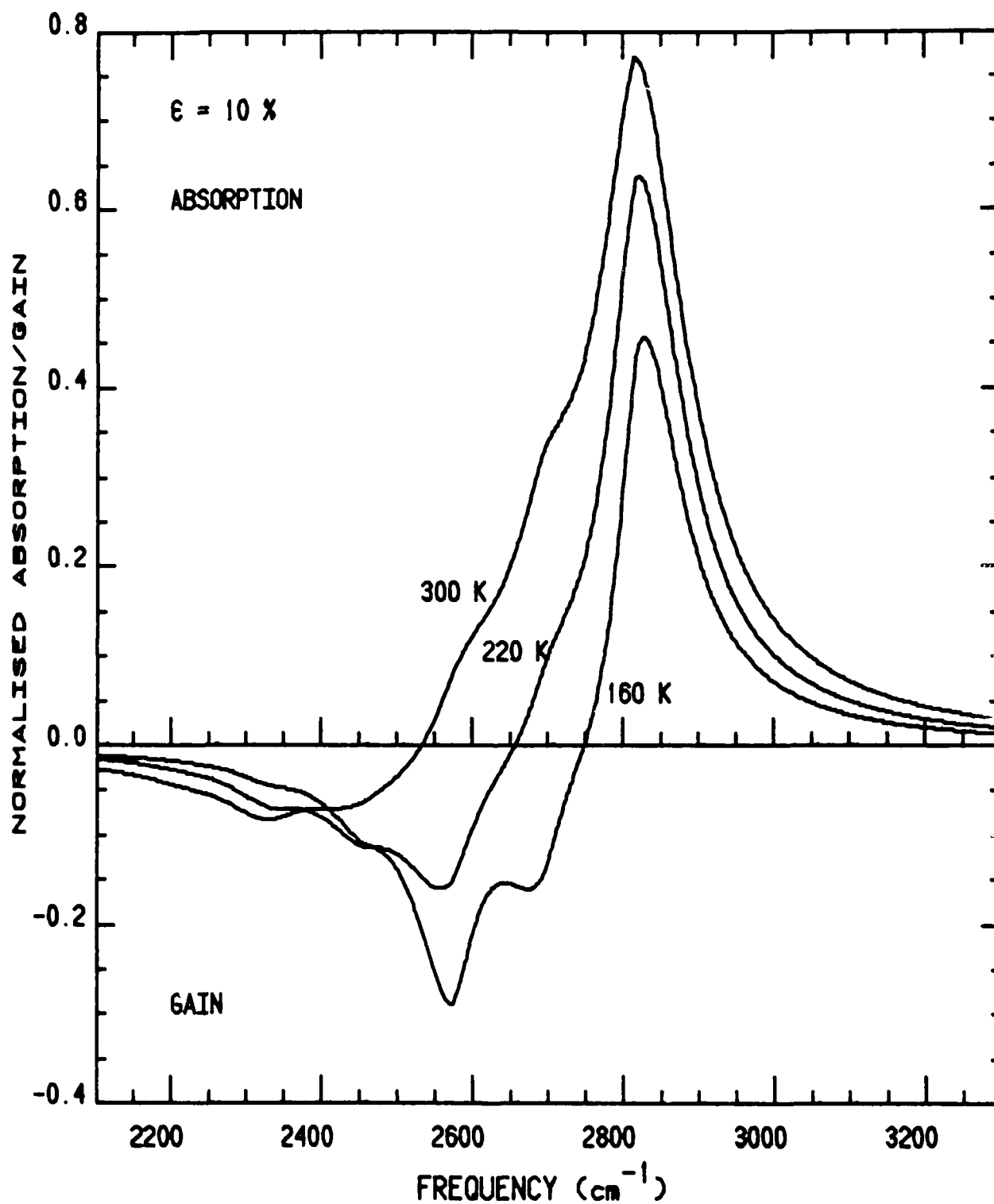


FIGURE 8. Normalised 0-branch absorption/gain of a 10%-excited D_2 mixture for the three values of temperature indicated.

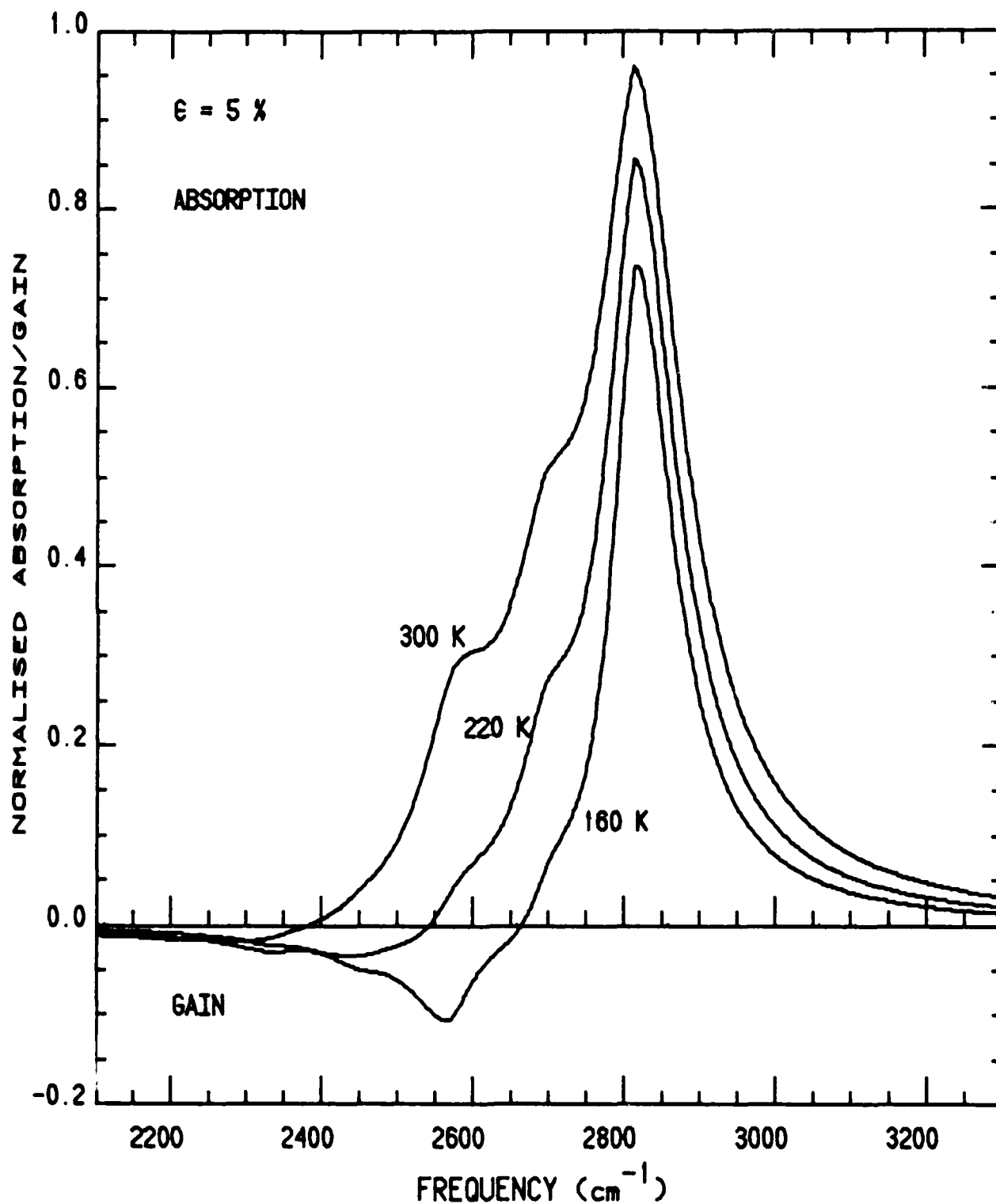


FIGURE 7. Normalised 0-branch absorption/gain of a 5%-Excited D_2 mixture for the three values of temperature indicated.

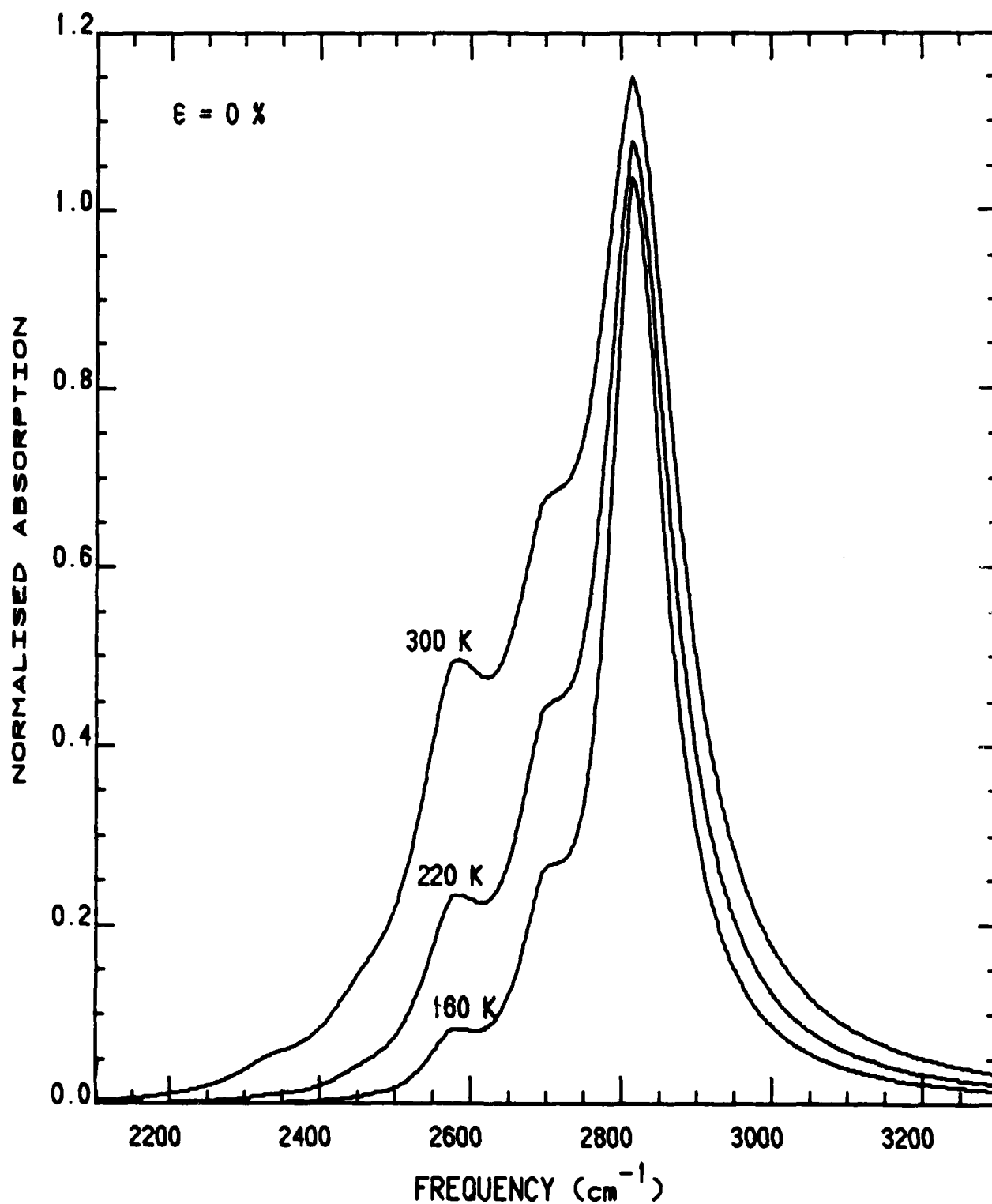


FIGURE 6. Normalised O-branch absorption of an unexcited D₂ molecule ($\epsilon = 0$) for the three values of temperature indicated.

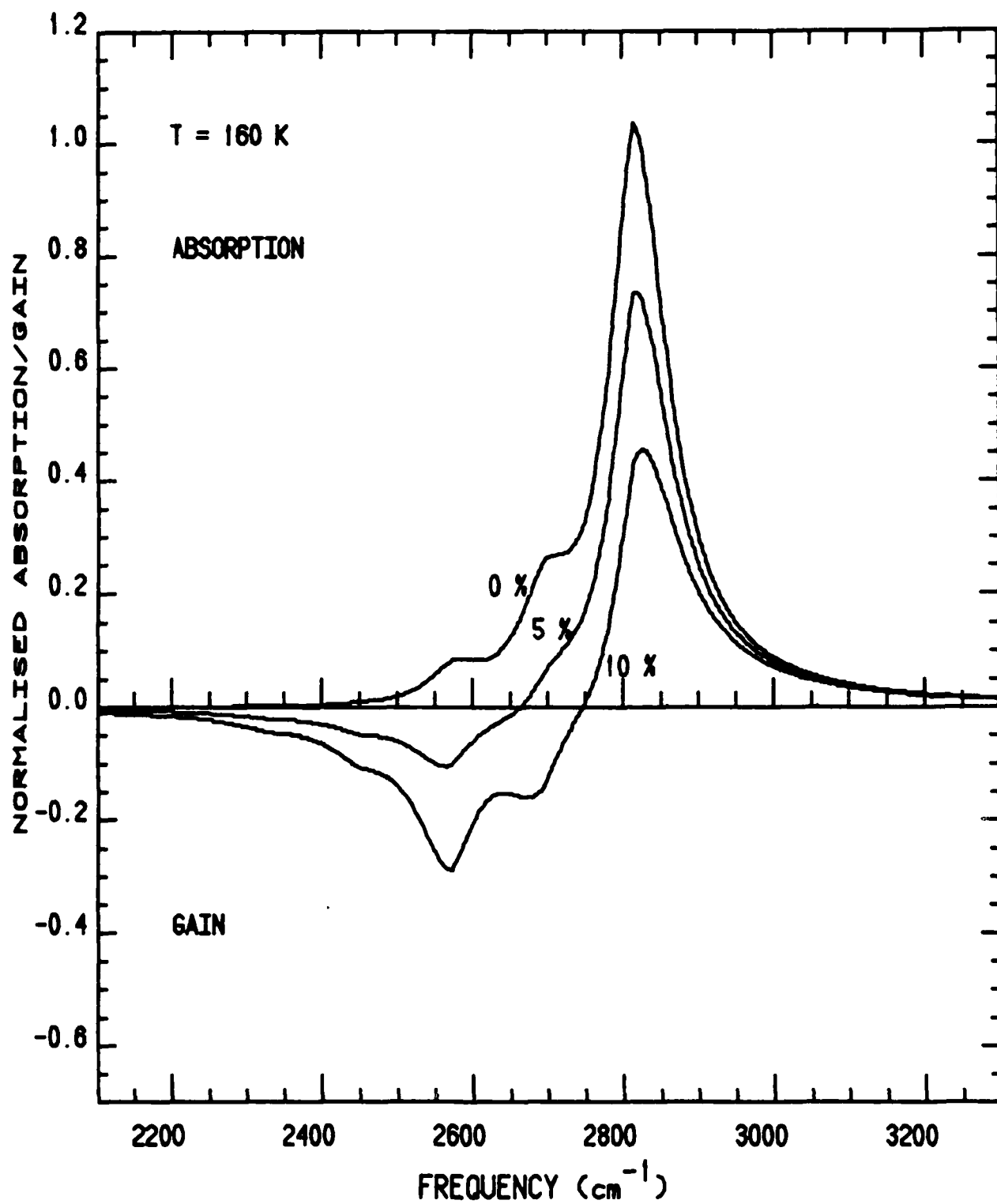


FIGURE 5. Normalised 0-branch absorption/gain of a D₂ mixture at 160 K as a function of frequency. The value of the excitation parameter ϵ is indicated on the curves.

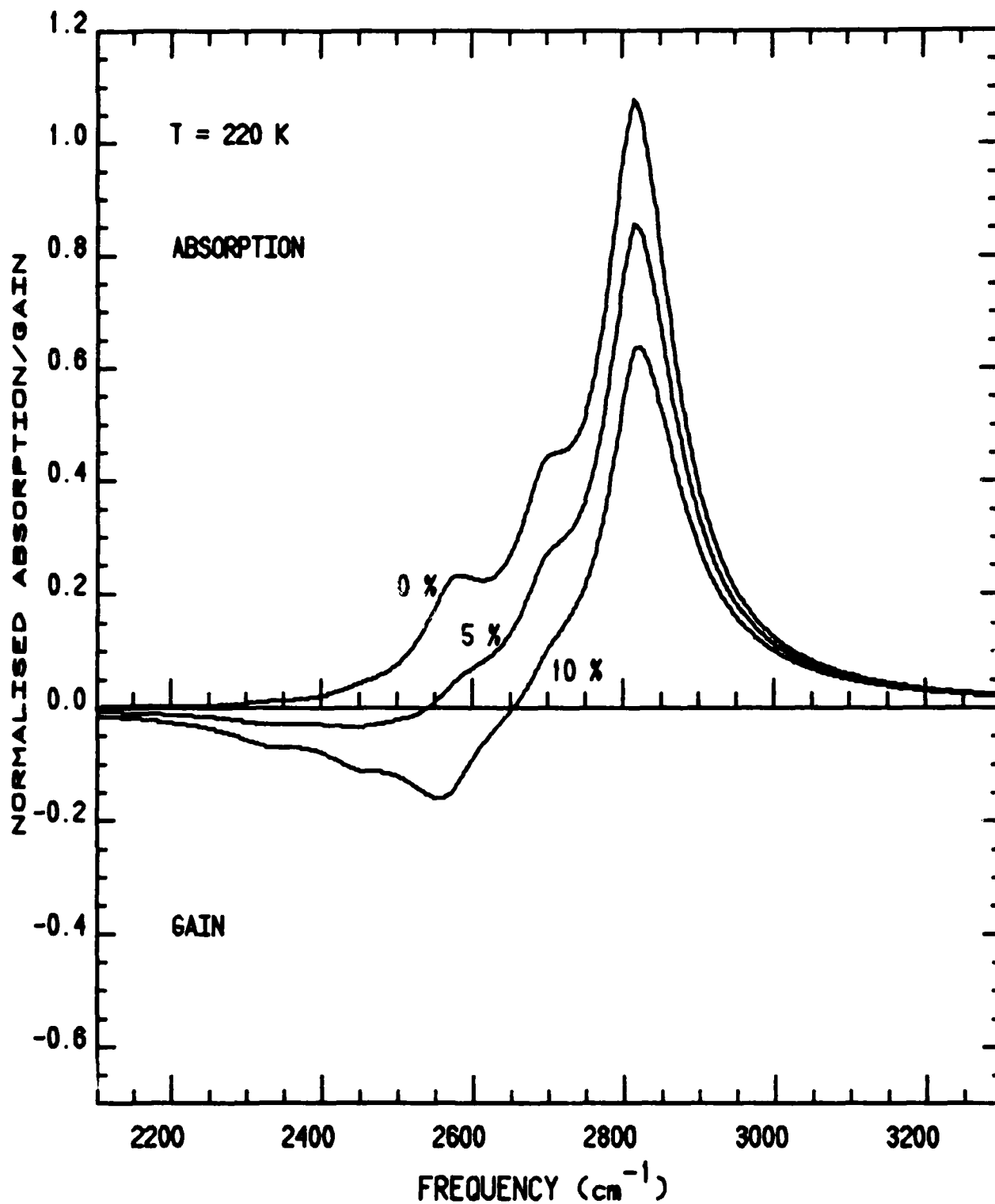


FIGURE 4. Normalised 0-branch absorption/gain of a D₂ mixture at 220 K as a function of frequency. The value of the excitation parameter ϵ is indicated on the curves.

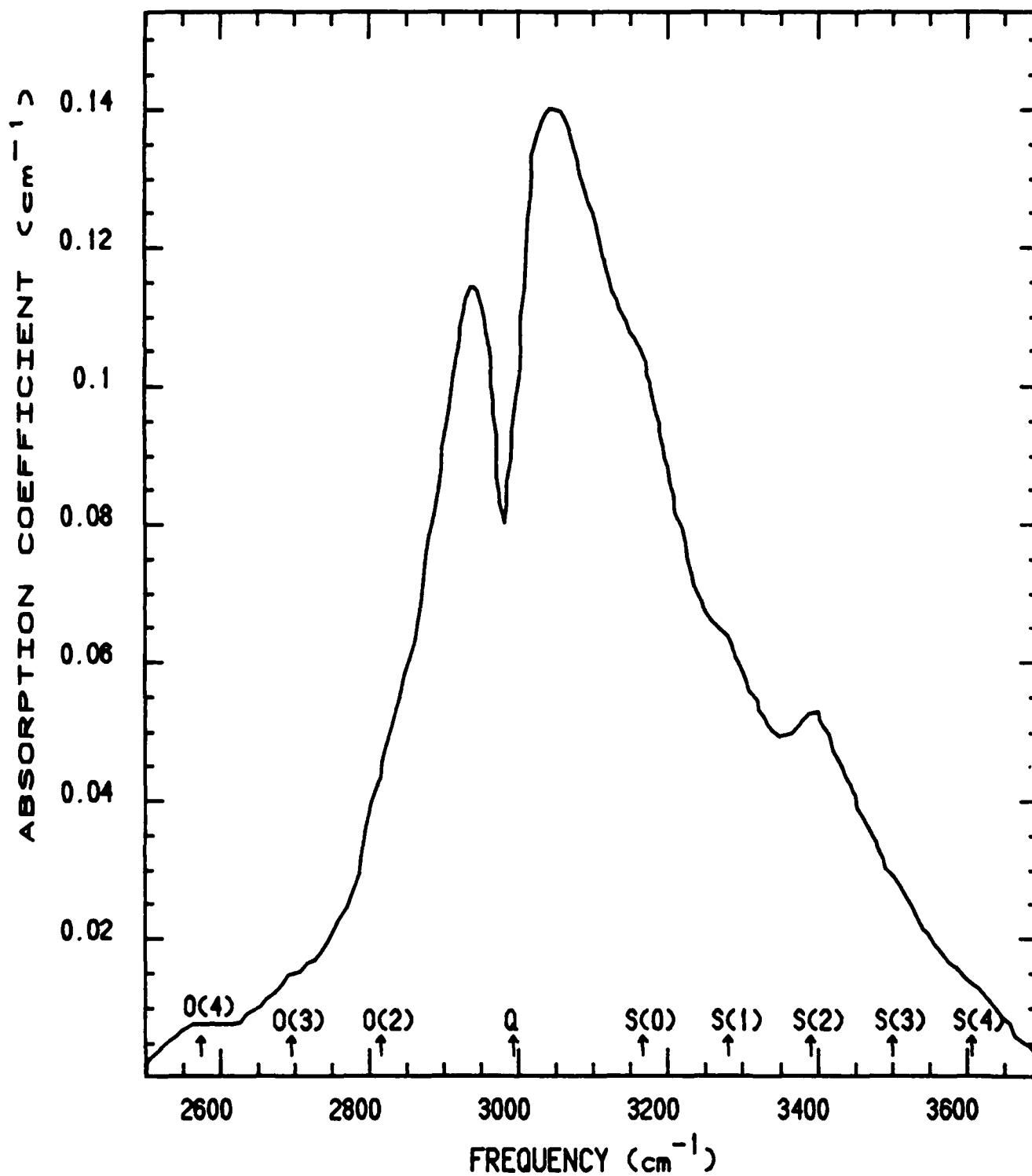


FIGURE 1. Absorption coefficient as a function of frequency for a D_2 :Ar mixture of 95:230 Amagat.

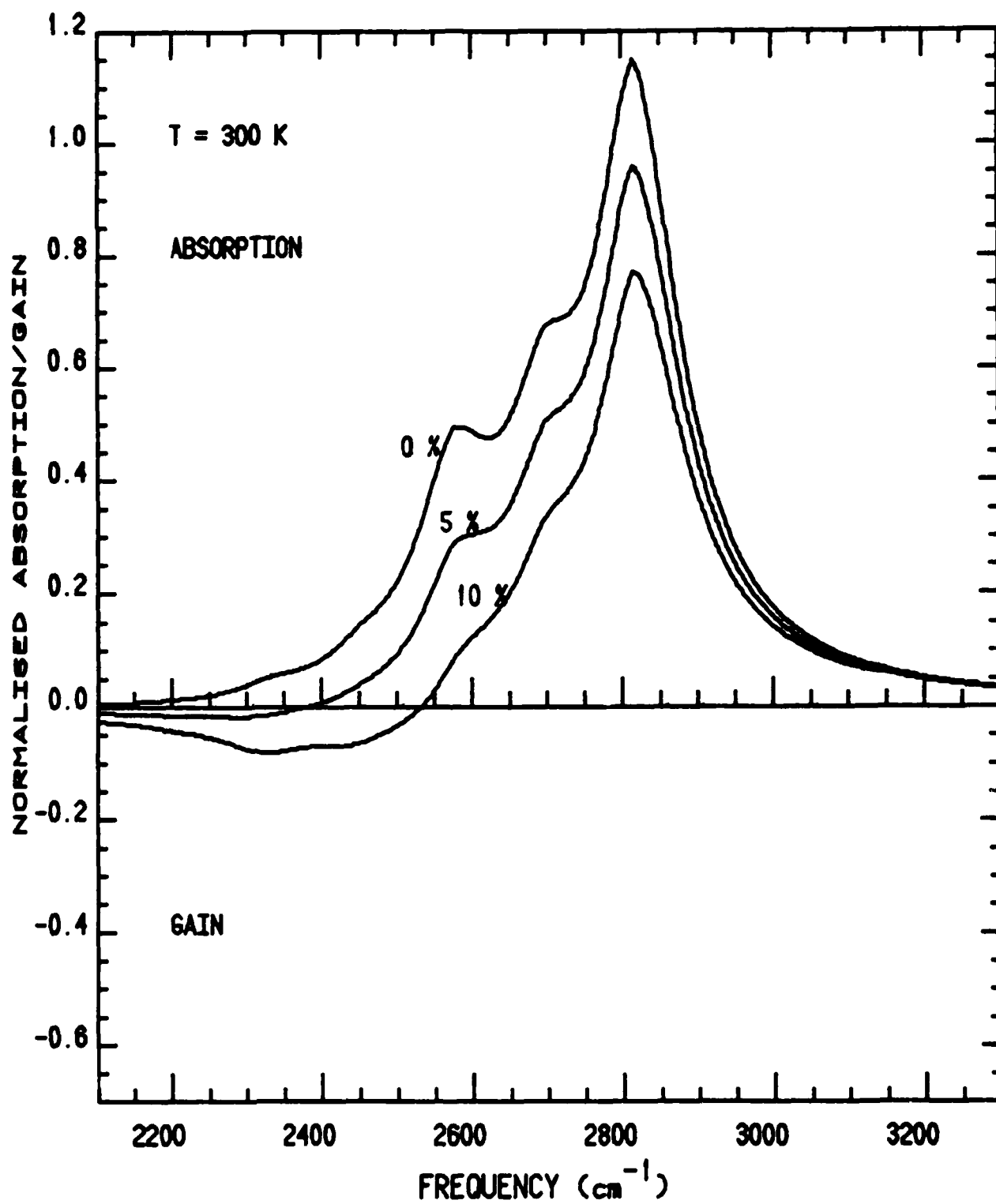


FIGURE 3. Normalised O-branch absorption/gain of a D_2 mixture at 300 K as a function of frequency. The value of the excitation parameter ϵ is indicated on the curves.

N.B. FIGURE 1 FOLLOWS FIGURE 3 IN ERROR.

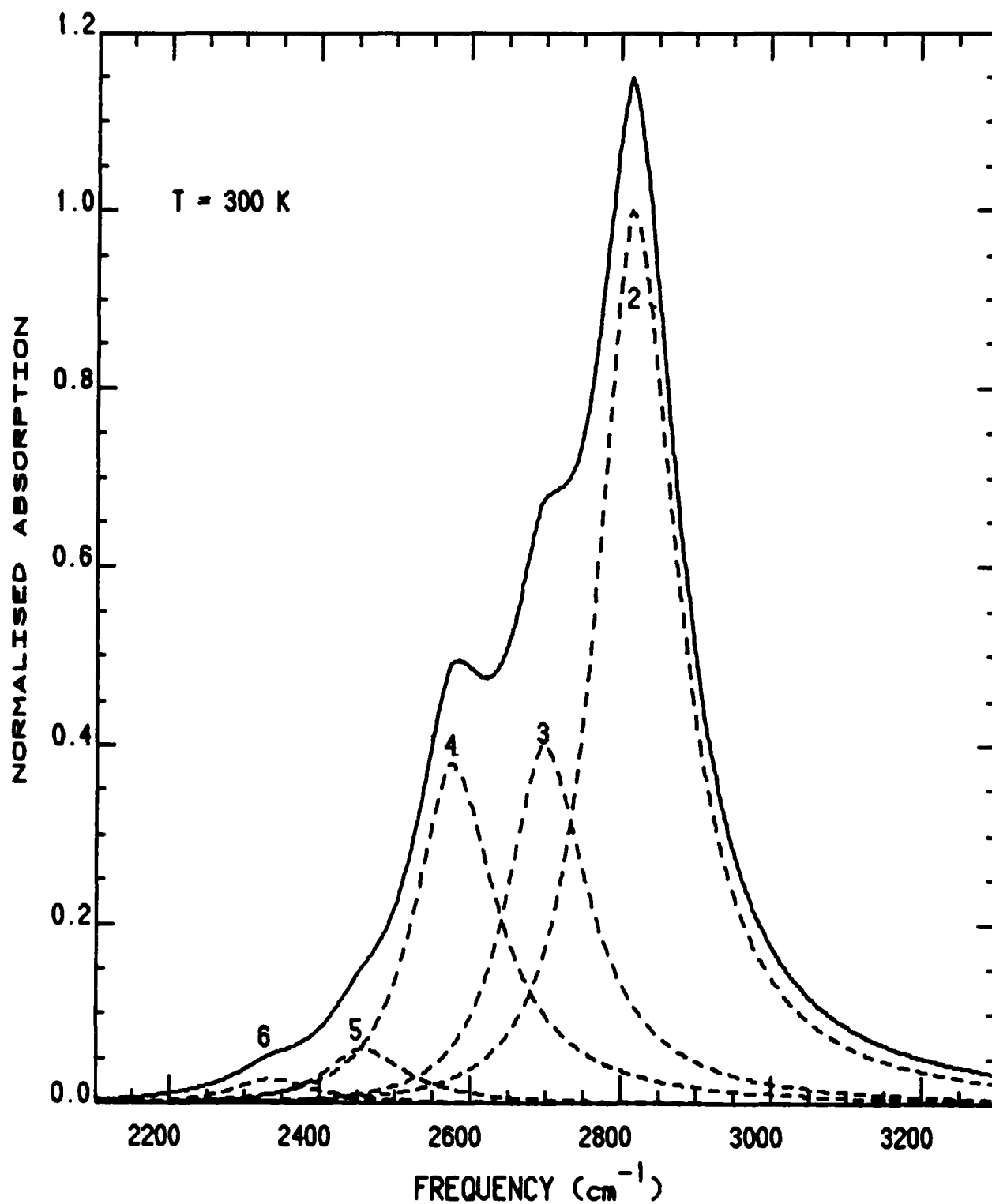


FIGURE 2. Normalised O-Branch absorption profile of an unexcited D_2 mixture ($\epsilon = 0$) at 300 K as a function of frequency. The individual O-branch transitions are shown as dashed curves.

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11. APPENDIX D

QUADRUPOLE MATRIX ELEMENTS $\langle vJ|Q|v'J' \rangle$

[11]: The quadrupole matrix elements are closely approximated by [10],

$$\langle vJ|Q|v'J' \rangle_{HD} = 0.192 - 3.05 \times 10^{-5} \bar{\nu}_J(HD)$$

$$\langle vJ|Q|v'J' \rangle_{D_2} = 0.172 - 3.28 \times 10^{-5} \bar{\nu}_J(D_2)$$

where $\bar{\nu}_J(HD)$ and $\bar{\nu}_J(D_2)$ are given for 0-branch transitions in Appendix B.

10. APPENDIX C

LINE SHAPE FUNCTIONS L_A , L_E

The line shape functions in absorption (L_A) and emission (L_E) are given by [2], [6]:

$$\begin{aligned}
 L_A(\nu) &= \{1 + (\Delta\nu/\delta q)^2\}^{-1} & \Delta\nu > 0 \\
 &= \{1 + (\Delta\nu/\delta q)^2\}^{-1} \exp(-hc\Delta\nu/kT) & \Delta\nu < 0 \\
 \\
 L_E(\nu) &= \{1 + (\Delta\nu/\delta q)^2\}^{-1} \exp(-hc\Delta\nu/kT) & \Delta\nu > 0 \\
 &= \{1 + (\Delta\nu/\delta q)^2\}^{-1} & \Delta\nu < 0
 \end{aligned}$$

where δq is the half width at half height and is given as a function of temperature by:

$$\delta q = \delta q_0 (T/300)^{1/2}$$

References [2] and [12] give δq_0 for collisions of HD with Ar and Kr as:

$$\begin{aligned}
 \delta q_0(\text{HD} + \text{Ar}) &= 78 \text{ cm}^{-1} \\
 \delta q_0(\text{HD} + \text{Kr}) &= 70 \text{ cm}^{-1}
 \end{aligned}$$

Values of δq_0 are not available for mixtures of D_2 with these buffer gases. However it is unlikely that they will be significantly different from the HD values. In addition, the results presented in this report are relatively insensitive to changes in the value of δq_0 . Thus a constant value, $\delta q_0 = 75 \text{ cm}^{-1}$, has been used throughout.

9. APPENDIX B

TABLE OF TRANSITION FREQUENCIES ($\bar{\nu}_J$)

Transition frequencies are derived from [5]:

Transition	$\bar{\nu}_J$ (HD) cm^{-1}	$\bar{\nu}_J$ (D ₂) cm^{-1}
0(2)	3365	2815
0(3)	3185	2694
0(4)	3005	2573
0(5)	2824	2451
0(6)	2645	2330
0(7)	2468	2209

The calculated values of the above functions are presented in the following tables.

For HD:

J or J'	T = 300K		T = 220K		T = 160K	
	$N_{VJ}/(Ng_J)$	$N_{V'J'}/(Ng_{J'})$	$N_{VJ}/(Ng_J)$	$N_{V'J'}/(Ng_{J'})$	$N_{VJ}/(Ng_J)$	$N_{V'J'}/(Ng_{J'})$
0	2.00×10^{-1}	1.91×10^{-1}	2.65×10^{-1}	2.55×10^{-1}	3.51×10^{-1}	3.38×10^{-1}
1	1.30×10^{-1}	1.27×10^{-1}	1.48×10^{-1}	1.46×10^{-1}	1.57×10^{-1}	1.56×10^{-1}
2	5.51×10^{-2}	5.59×10^{-2}	4.58×10^{-2}	4.75×10^{-2}	3.14×10^{-2}	3.36×10^{-2}
3	1.52×10^{-2}	1.63×10^{-2}	7.93×10^{-3}	8.87×10^{-3}	2.82×10^{-3}	3.34×10^{-3}
4	2.74×10^{-3}	3.16×10^{-3}	7.65×10^{-4}	9.47×10^{-4}	1.13×10^{-4}	1.54×10^{-4}
5	3.21×10^{-4}	4.07×10^{-4}	4.11×10^{-5}	5.77×10^{-5}	2.03×10^{-6}	3.29×10^{-6}
6	2.45×10^{-5}	3.47×10^{-5}	1.23×10^{-6}	2.01×10^{-6}	1.63×10^{-8}	3.26×10^{-8}
7	1.22×10^{-6}	1.96×10^{-6}	2.05×10^{-8}	4.01×10^{-8}	5.86×10^{-11}	1.49×10^{-10}

For D₂:

J or J'	T = 300K		T = 220K		T = 160K	
	$N_{VJ}/(Ng_J)$	$N_{V'J'}/(Ng_{J'})$	$N_{VJ}/(Ng_J)$	$N_{V'J'}/(Ng_{J'})$	$N_{VJ}/(Ng_J)$	$N_{V'J'}/(Ng_{J'})$
0	1.82×10^{-1}	1.76×10^{-1}	2.44×10^{-1}	2.36×10^{-1}	3.28×10^{-1}	3.17×10^{-1}
1	6.84×10^{-2}	6.68×10^{-2}	8.26×10^{-2}	8.10×10^{-2}	9.56×10^{-2}	9.43×10^{-2}
2	7.71×10^{-2}	7.68×10^{-2}	7.55×10^{-2}	7.61×10^{-2}	6.52×10^{-2}	6.68×10^{-2}
3	1.63×10^{-2}	1.67×10^{-2}	1.17×10^{-2}	1.23×10^{-2}	6.48×10^{-3}	7.03×10^{-3}
4	1.03×10^{-2}	1.11×10^{-2}	4.87×10^{-3}	5.41×10^{-3}	1.51×10^{-3}	1.76×10^{-4}
5	1.23×10^{-3}	1.38×10^{-3}	3.44×10^{-4}	4.09×10^{-4}	5.10×10^{-5}	6.57×10^{-5}
6	4.39×10^{-4}	5.25×10^{-4}	6.57×10^{-5}	8.50×10^{-5}	4.04×10^{-6}	5.83×10^{-6}
7	2.94×10^{-5}	3.78×10^{-5}	2.12×10^{-6}	3.02×10^{-6}	4.66×10^{-8}	7.69×10^{-8}

8. APPENDIX A

EVALUATION OF $N_{vJ}/(Ng_J)$ AND $N_{v'J'}/(Ng_{J'})$

$N_{vJ}/(Ng_J)$ and $N_{v'J'}/(Ng_{J'})$ are evaluated for $v = 0$ and $v' = 1$.

From [9]:

$$\frac{N_{vJ}}{Ng_J} = \frac{g_T \exp[-J(J+1)hcB/kT]}{\sum_J g_T g_J \exp[-J(J+1)hcB/kT]}$$

$$\frac{N_{v'J'}}{Ng_{J'}} = \frac{g_T \exp[-J'(J'+1)hcB'/kT]}{\sum_{J'} g_T g_{J'} \exp[-J'(J'+1)hcB'/kT]}$$

For HD [5], [9]:

$$g_T = 1 \text{ (all } J), B = 44.67 \text{ cm}^{-1}, B' = 42.74 \text{ cm}^{-1}$$

For D₂ [5], [9]:

$$g_T = 6 \text{ (} J \text{ even)} = 3 \text{ (} J \text{ odd)}, B = 29.91 \text{ cm}^{-1}, B' = 28.85 \text{ cm}^{-1}$$

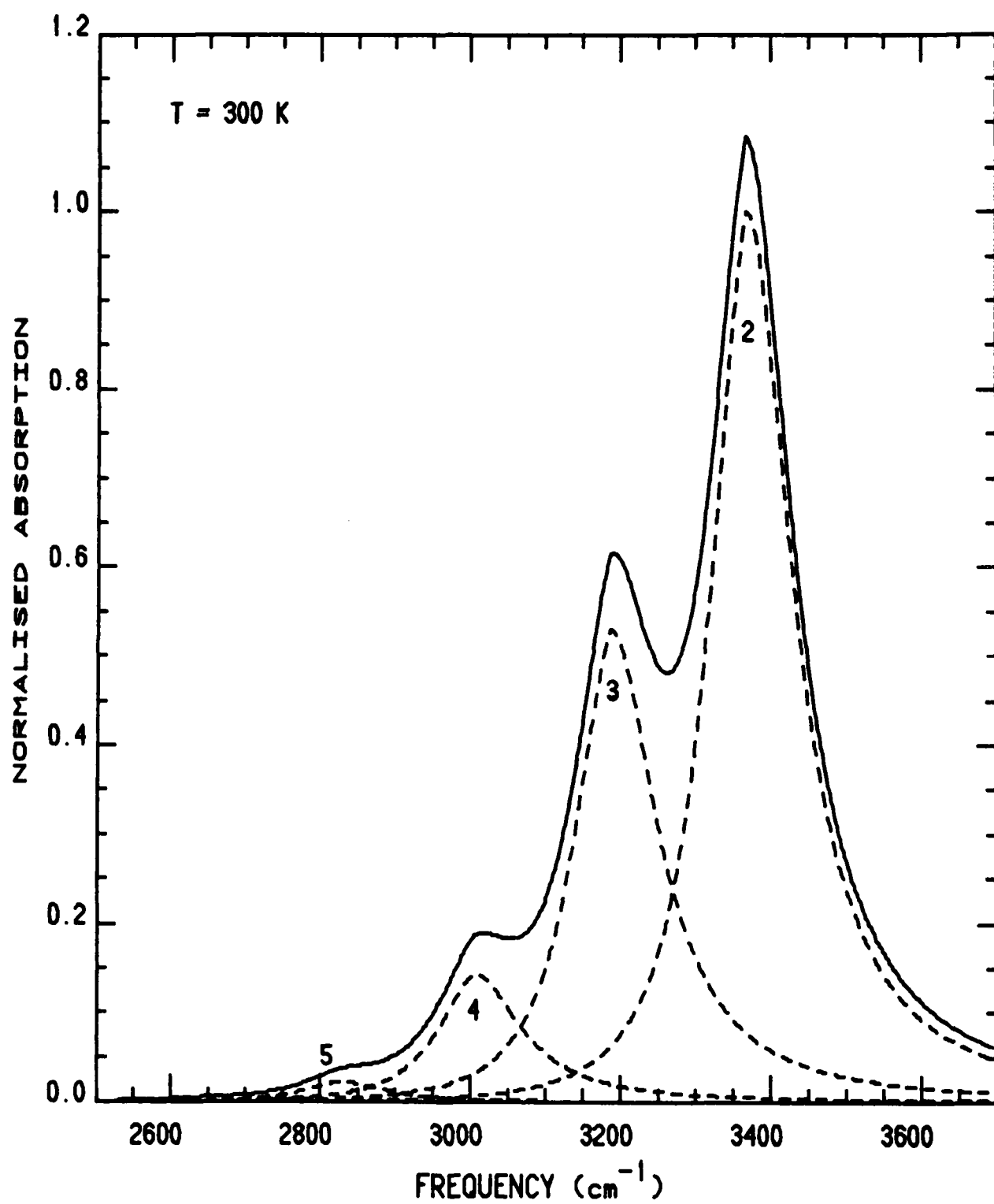


FIGURE 9. Normalised O-branch absorption profile of an unexcited HD mixture ($\epsilon = 0$) at 300 K as a function of frequency. The individual O-branch transitions are shown as dashed curves.

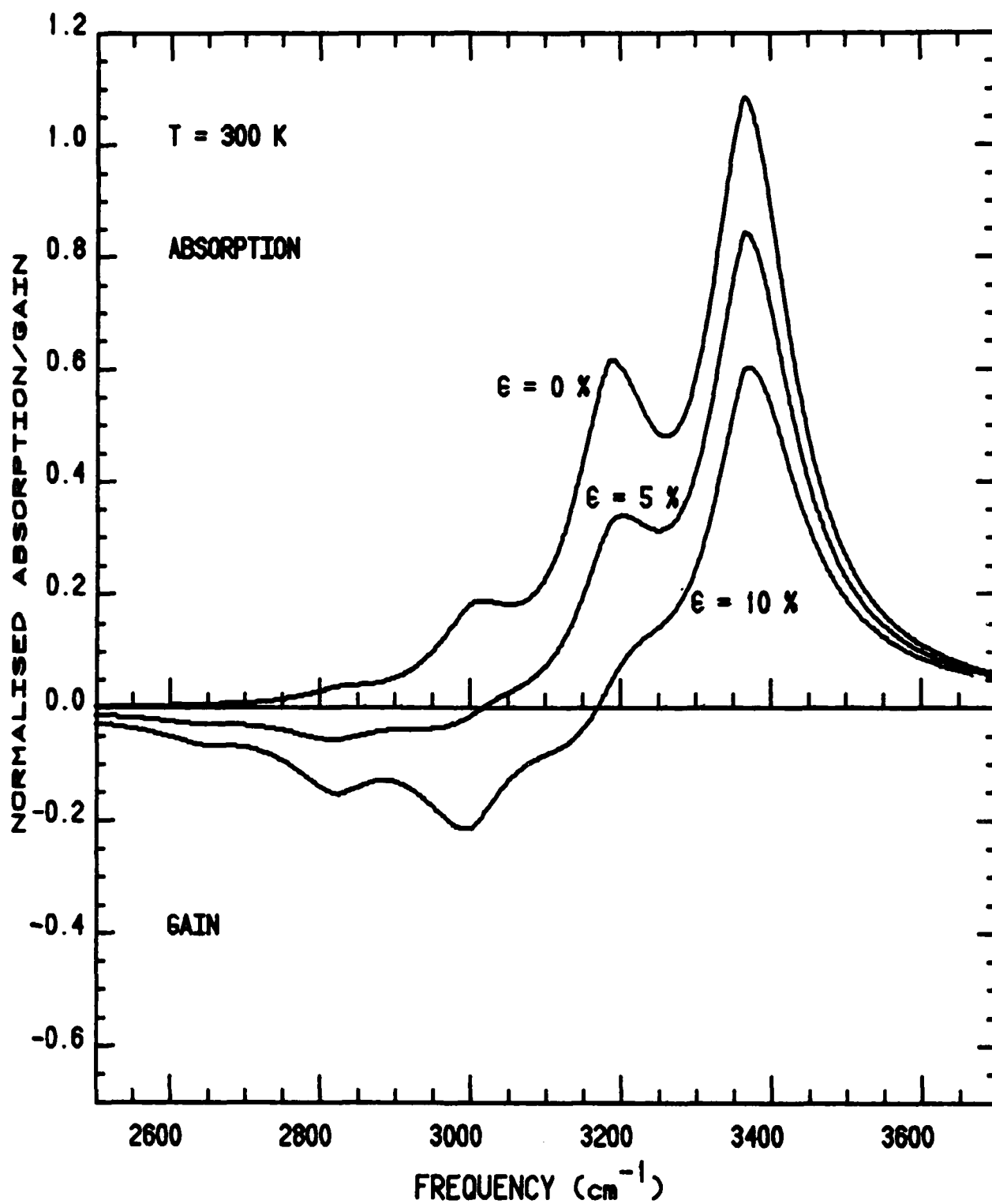


FIGURE 10. Normalised 0-branch absorption/gain of a HD mixture at 300 K as a function of frequency. The value of the excitation parameter ϵ is indicated on the curves.

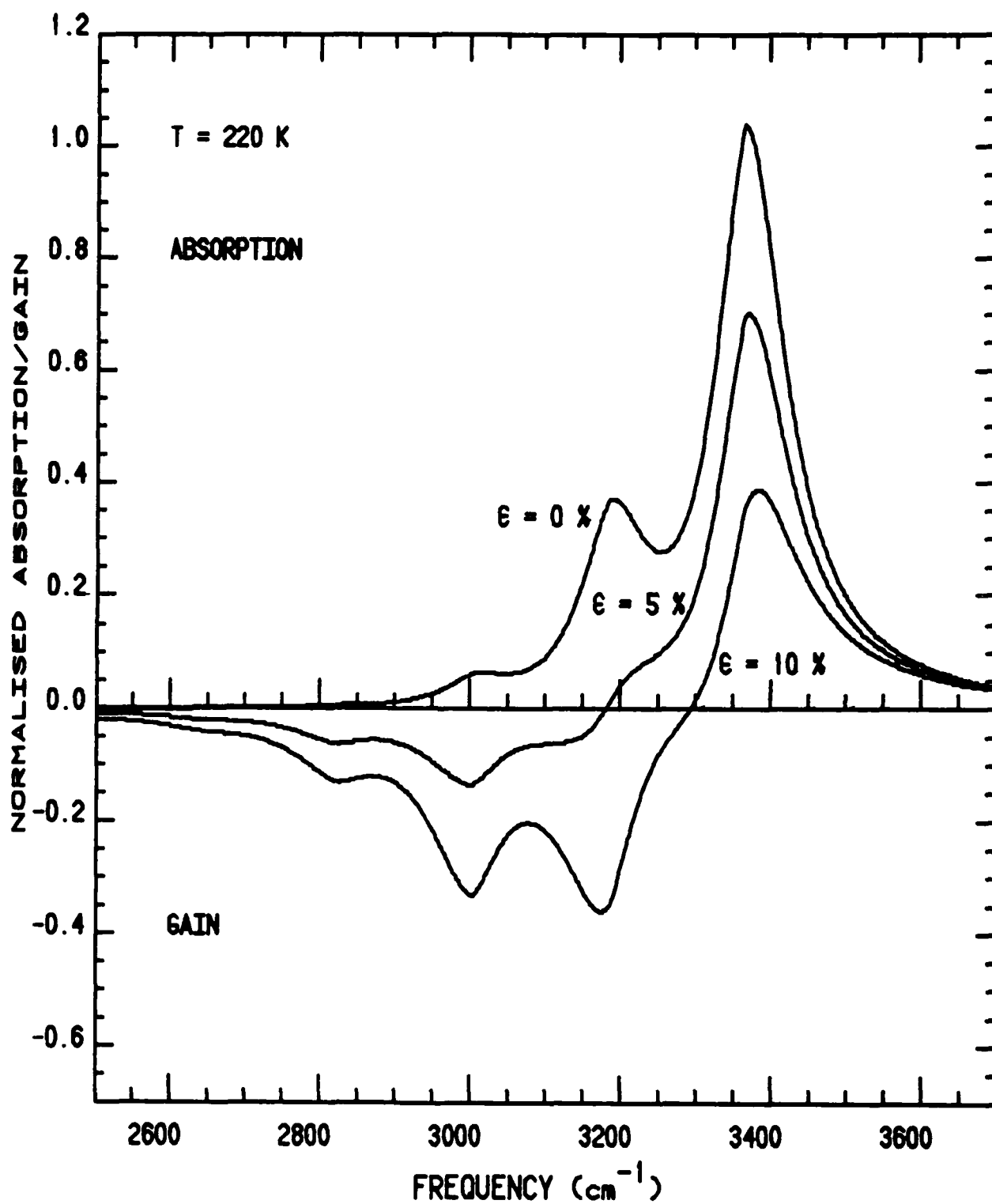


FIGURE 11. Normalised 0-branch absorption/gain of a HD mixture at 220 K as a function of frequency. The value of the excitation parameter ϵ is indicated on the curves.

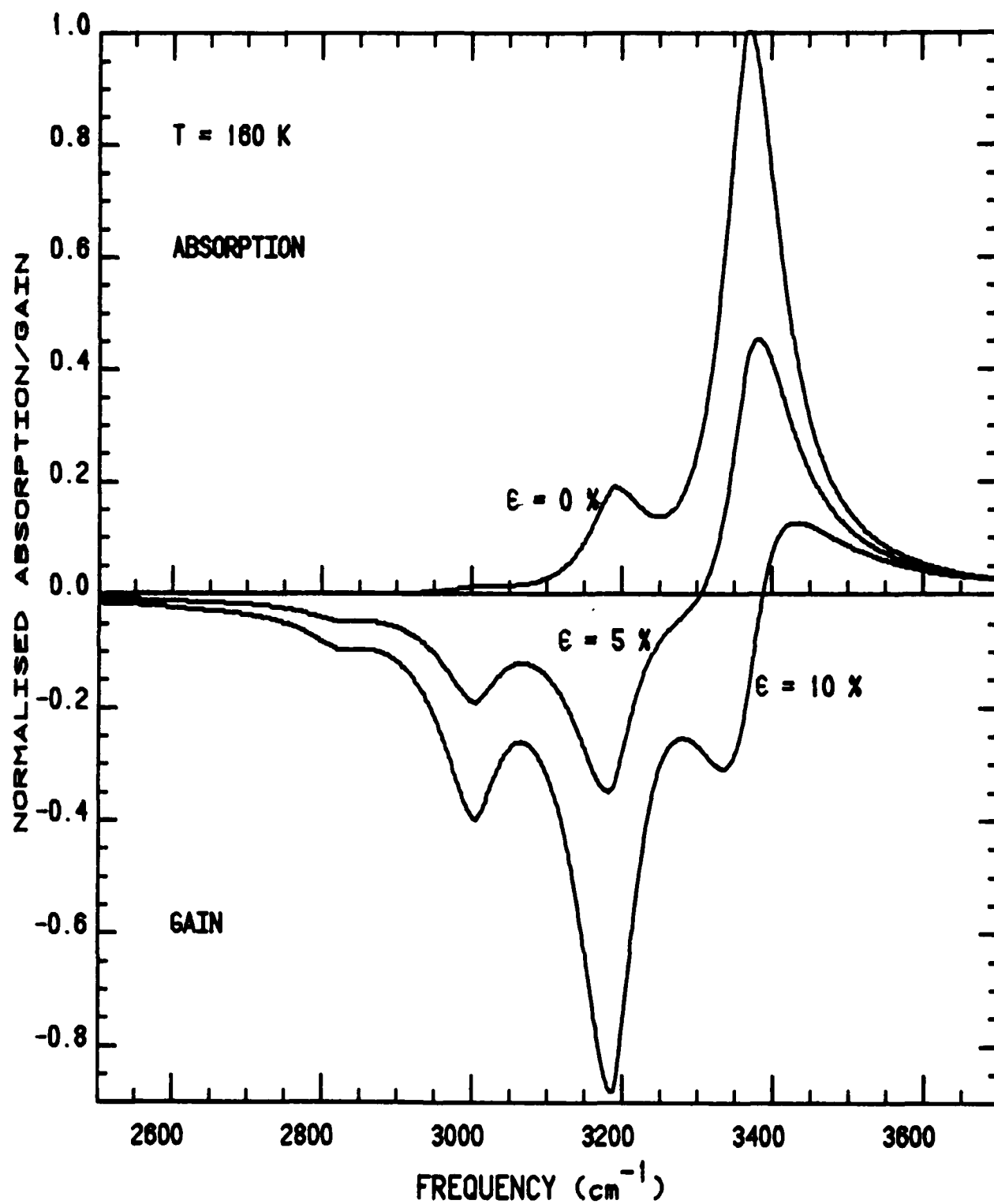


FIGURE 12. Normalised 0-branch absorption/gain of a HD mixture at 160 K as a function of frequency. The value of the excitation parameter ϵ is indicated on the curves.

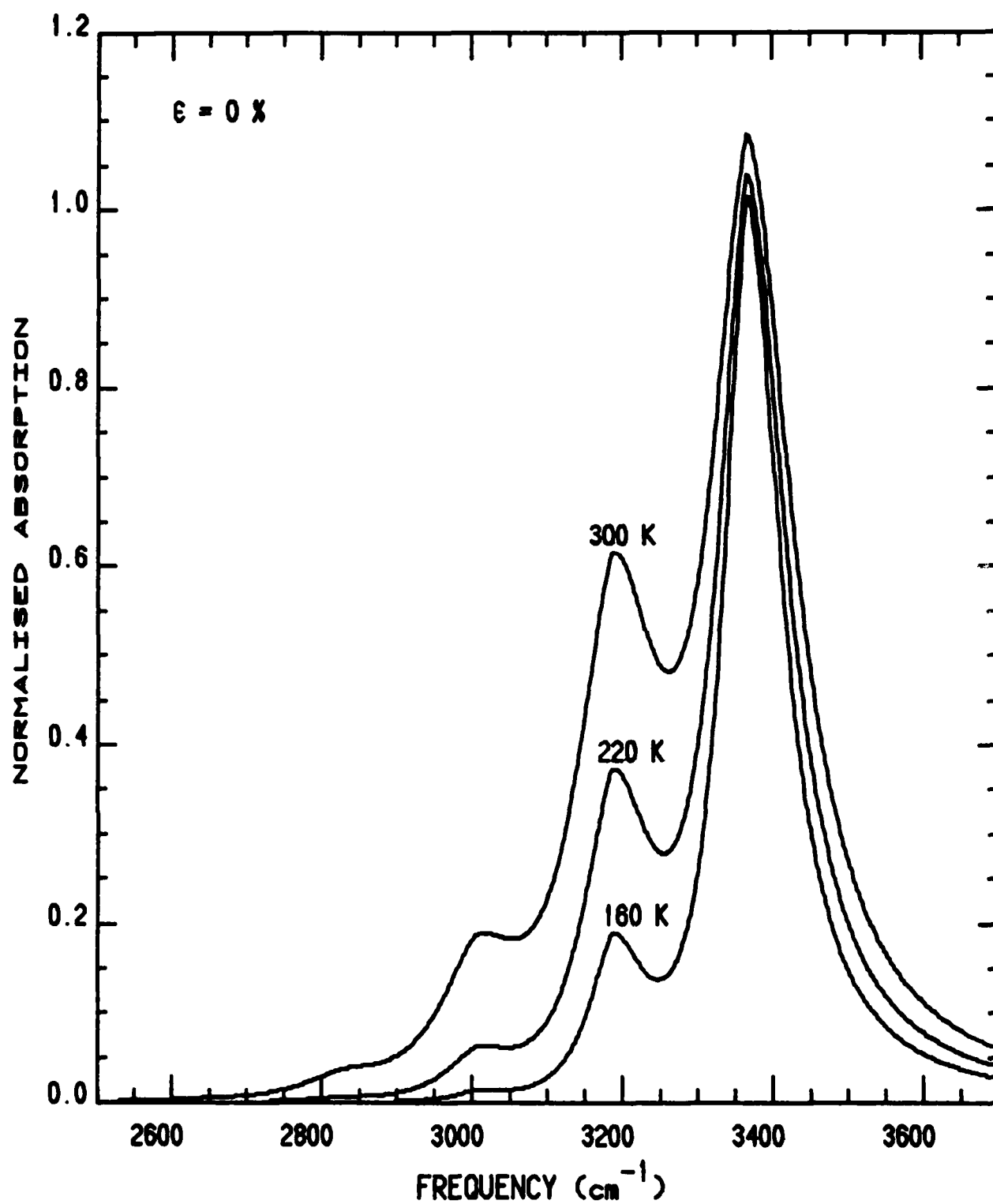


FIGURE 13. Normalised O-branch absorption of an unexcited HD mixture ($\epsilon = 0$) for the three values of temperature indicated.

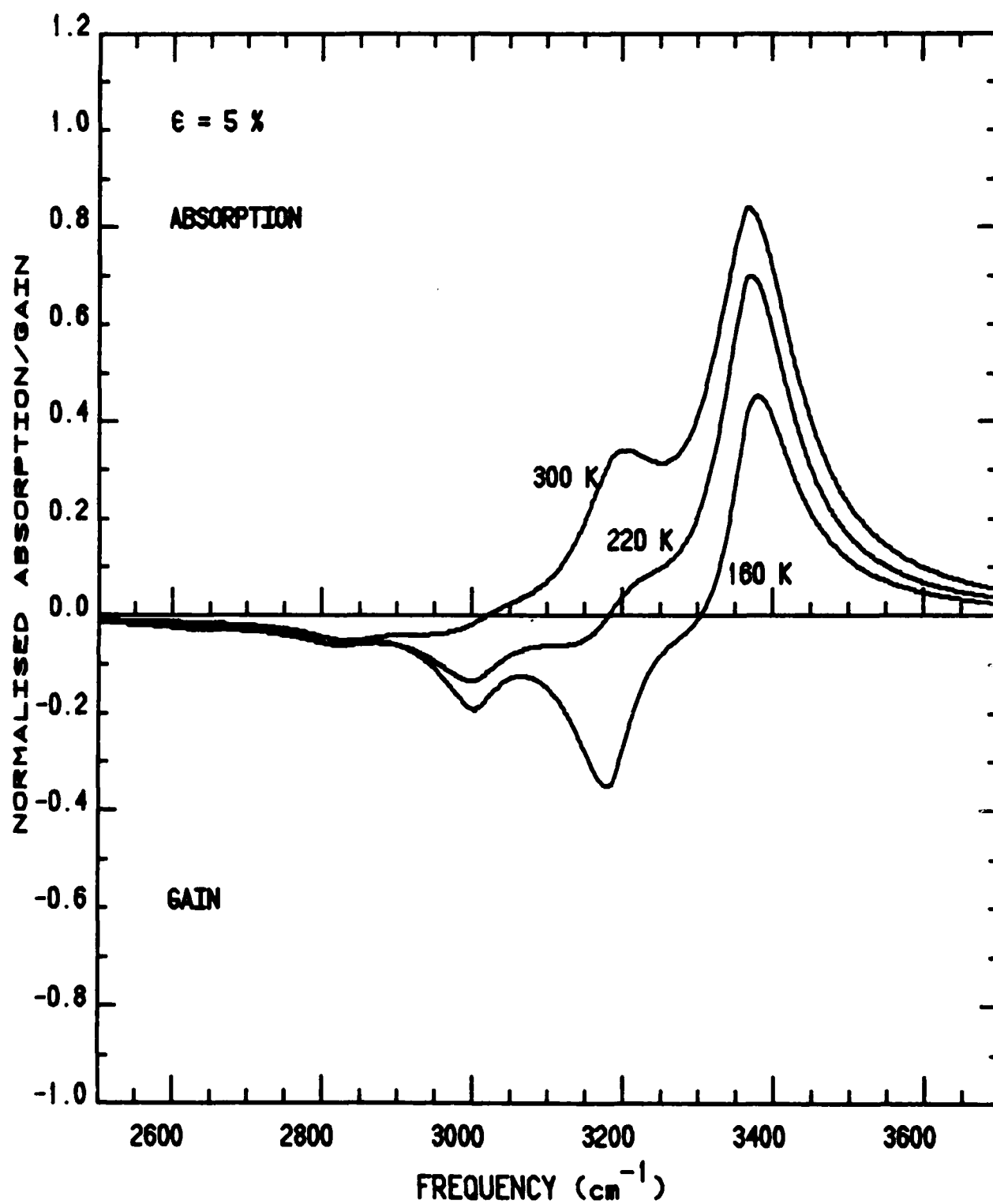


FIGURE 14. Normalised O-branch absorption/gain of a 5%-excited HD mixture for the three values of temperature indicated.

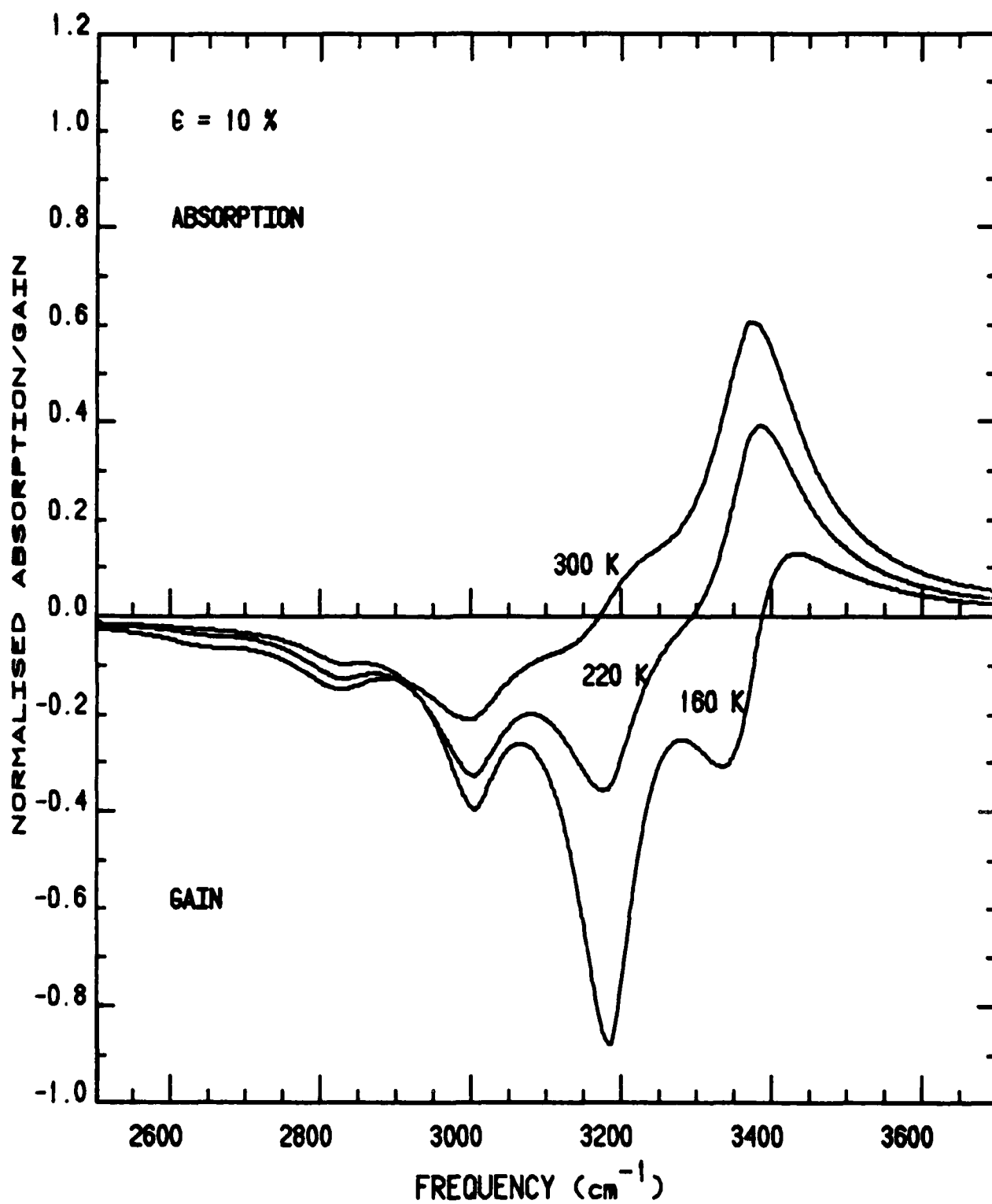


FIGURE 15. Normalised O-branch absorption/gain of a 10%-excited HD mixture for the three values of temperature indicated.

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